The incommensurate charge-density-wave instability in the extended three-band Hubbard model

F. Becca, F. Bucci, and M. Grilli
Istituto Nazionale di Fisica della Materia and Dipartimento di Fisica, Università di Roma "La Sapienza", Piazzale A. Moro 2, Roma, Italy 00185

The infinite-U three-band Hubbard model is considered in order to describe the CuO_2 planes of the high temperature superconducting cuprates. The charge instabilities are investigated when the model is extended with a nearest-neighbor repulsion between holes on copper d and oxygen p orbitals and in the presence of a long-range Coulombic repulsion. It is found that a first-order valence instability line ending with a critical point is present like in the previously investigated model without long-range forces. However, the dominant critical instability is the formation of incommensurate charge-density-waves, which always occur before the valence-instability critical point is reached. An effective singular attraction arises in the proximity of the charge-density wave instability, accounting for both a strong pairing mechanism and for the anomalous normal state properties.

I. INTRODUCTION

The crucial structural elements, present in all the superconducting cuprates, are the CuO_2 planes. At half-filling, i.e. one hole per unit cell in the CuO_2 planes, these materials are antiferromagnetic charge-transfer insulators and the holes mostly occupy the copper sites. The holes added by doping reside on the CuO_2 planes and have a large amplitude on oxigen, thus showing that the strong hole-hole repulsion on copper is a relevant feature. Upon doping the system is driven toward a paramagnetic metallic phase which becomes superconducting at low temperature. The metallic phase above T_c presents many anomalous features, which contrast with the usual behavior of the normal Fermi liquids.

The failure of Landau Fermi-liquid theory in the metallic phase of the cuprates has been ascribed to singular interactions arising in the proximity of some critical point at zero temperature (quantum critical point, QCP) [1–6]. More specifically the complex behavior of these systems was recently interpreted in terms of a proximity to an incommensurate charge-density-wave (ICDW) transition [5–8] located at zero temperature near the optimal doping. In the quantum critical region above it no energy scale besides temperature rules the physics and strong critical fluctuations are responsible for both non-Fermi liquid behavior and large superconducting critical temperatures. As soon as superconductivity takes place the CDW instability is hindered and can only be recovered by destroying the superconducting coherent state, like, e.g., in transport experiments under strong magnetic fields [9]. In the underdoped compounds, instead the instability would occur at finite temperature were it not for the quenching due to superconducting local fluctuations which can give rise to the appearance of charge and spin gaps of d-wave symmetry as experimentally found [10–14] at a temperature T^* above T_c. According to this proposal [7,8], the underlying hindered charge instability provides the temperature dependent pairing potential needed to explain the high crossover temperature T^* of the gap formation and the peculiar doping dependence of T^* (strongly increasing with decreasing doping, [10–12]) with respect to the value of the charge gap at T=0 (nearly constant with decreasing doping, [11,15]).

The existence of a QCP near optimal doping is supported by several experimental findings. In particular recent transport measurements in $La_{2-x}Sr_xCuO_4$ (LSCO) under strong magnetic field [9] investigated the normal phase of these systems when superconductivity is suppressed. This analysis shows the existance of a QCP near optimal doping. Indications in this sense are also provided by neutron scattering [16] and by the qualitative changes of behavior at optimal doping detected by optical spectroscopy [17], NMR [14], susceptibility [18], neutron scattering [13], photoemission [10–12], specific heat [15], thermoelectric power [19], Hall coefficient [20], resistivity [9,18,21]. It is also suggestive that several quantities (resistivity, Hall number, uniform susceptibility) display a scaling behavior with a typical energy scale, which vanishes at optimal doping [22–24].

Many indications exist that the above QCP involves charge ordering. Generically, the sizable doping at which the QCP occurs suggests that charge degrees of freedom are substantially involved in the ordering phenomenon. A direct observation of charge-driven ordering was possible by neutron scattering [25–27], in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ where the related Bragg peaks were detected. For this specific compound the low temperature tetragonal lattice structure pins the CDW and gives static order and semiconducting behavior (see also the case of $La_{1.88}Ba_{0.12}CuO_4$). Increasing the

Sr content at fixed Nd concentration, the pinning effect is reduced leading to metallic and superconducting behavior. In this latter case, the existence of dynamical ICDW fluctuations is suggested by the presence of dynamical incommensurate spin scattering, although the charge peaks are too weak to be observed. In this regard, also the $La_{2-x}Sr_xCuO_4$ is expected to display dynamical charge fluctuations with doping-dependent spatial modulation as indeed observed in the magnetic scattering [28]. ICDW have been proposed from extended X-ray absorption fine structure (EXAFS) experiments both in optimally doped LSCO [29] and $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212) [30]. Superstructures have also been detected in Bi-2212 from X-ray diffraction [31].

The occurrence of an ICDW-QCP is theoretically substantiated by explicit findings within the single-band infinite-U Hubbard model in the presence of an Holstein electron-phonon coupling. In the absence of long-range (LR) coulombic forces and for sizable but realistic electron-phonon coupling, this model displays phase separation (PS) into macroscopically large half-filled insulating regions and metallic hole-rich doped regions. When the LR Coulomb repulsion between holes is included the $q = 0$ charge instability is prevented. Nevertheless a phase-separation tendency remains in the system and shows up on a local basis: The PS region of the phase diagram is replaced at small doping by a smaller ICDW instability region characterized by a non-vanishing incommensurate wavevector q_c.

As discussed above, the occurrence of an incommensurate charge instability and of the related singular attractive interactions is a key point, which could provide a unified explanation for both the normal and superconducting properties of the cuprates. In this regard it seems quite important to investigate how generic and robust is the above scenario based on an ICDW instability arising from the competition between PS and LR repulsion.

As far as PS is concerned, this is a common feature of strongly correlated electron systems: It has been found in models with nearest-neighbour Coulomb interactions [32–36], in magnetic models [37,38], and in models with phononic interactions [39,40]. Indeed a strong on-site correlation drastically renormalizes the kinetic energy, which would tend to delocalize the carriers into Bloch quasiparticle states. Then short-range interactions (magnetic, phononic, nearest-neighbor coulombic...) introducing effective attractions between the carriers may dominate and give rise to charge aggregation in highly doped metallic regions together with charge depletion in spatially separated (ordered) regions with no itinerant charges.

As pointed out by Emery and Kivelson [41] LR Coulomb forces effectively oppose the separation of charged particles suppressing long-wavelenght density fluctuations. This may lead to either dynamical slow density fluctuations [41] or static ICDW [5], [6], [34], [42]. However, whereas the occurrence of PS is well established in many models of strongly interacting electrons, the occurrence of ICDW from LR coulombic repulsion frustrating PS in a microscopic electron model was only discussed in a single-band Hubbard-Holstein model [5,39] and only marginally considered in a three-band extended Hubbard model [34]. Then, although the physical ingredients seem fully general, to gain further understanding of the ICDW-QCP scenario, it is desirable to investigate other theoretical models of strongly interacting electrons in the presence of LR forces. In this way one can both (a) explicitly check the generic character of the physical mechanism and (b) highlight the specific features arising from the various interactions. In this regard, the simple single-band Hubbard-Holstein model having already been considered, the natural extension seems to be a multiband Hubbard model with purely electronic interactions.

The modelization of CuO_2 planes by the three-band Hubbard model, in which the copper and oxigen degrees of freedom are explicitly taken into account, was proposed by Varma *et al.* together with the inclusion of the nearest-neighbor Coulomb repulsion in order to stress the role of the charge-transfer (copper-oxigen) fluctuations [43]. Littlewood *et al.* [44], [45], using a weak-coupling approach (for a finite not too large repulsion on copper site) have found that the presence of a nearest-neighbour Coulomb repulsion V of order of the bandwidth can drive the system toward a valence instability (VI), characterized by strong charge fluctuations between the copper d orbitals and the oxigen p orbitals. If the total number of particles is allowed to fluctuate, the VI has been shown [32] to be embedded in an unstable region where the system phase-separates. Also in a strong-coupling approach [33], [34] the mean-field solution at fixed number of particle displays a VI line ending at the so-called VI point, where the charge-transfer excitonic mode completely softens. However, when the number of particles can fluctuate, this line is surrounded by a region of negative compressibility, i.e. by a region in which the system phase-separates. Therefore PS always occurs both in weak and strong coupling before any VI takes place.

In this paper we address the problem of introducing the LR forces in a three-band Hubbard model. In this way we study how the charge-transfer fluctuations, which are able to provide a strong attraction in the particle-hole channel and lead to PS in the pure short-range case, are affected by a Coulomb potential. In section II we introduce the model. In particular we work within a strong coupling approach: An infinite repulsion between holes on the same copper site is handled by means of a standard slave-boson technique. Fluctuations around mean-field values are taken into account up to the gaussian order using a systematic $1/N$ expansion. This section is rather technical and the uninterested reader can skip it only retaining the form of the model provided by the pure short range terms [Eq. (1) with $U_d = \infty$] and by the LR potential [Eqs. (7)-(9)].

In section III we will show that, as in the LR Hubbard-Holstein model, the phase diagram of three-band Hubbard model in the presence of LR interactions displays a finite-q instability related to CDW formation. We find a singular scattering between quasiparticles near the line on which the generalized density-density correlation functions diverge at a finite q_c. By contrast in the proximity of the VI line no singular scattering is found. Only at the isolated VI point the interaction takes a singular form due to vanishing of the excitonic energy, but this has no physically relevant consequences since the VI point is always inside the unstable region with respect to the ICDW formation. This conclusion differs from the results obtained by Varma in a three-band Hubbard model [3,4], where an instability related to intracell charge-transfer currents was found, together with singular interactions. However, in the model considered by Varma a relevant role was played by general symmetry properties, which are absent in our case, thus rendering a direct comparison between the two models and results questionable. In section IV we discuss the effective interaction between quasiparticles, whereas in section V we draw our conclusions.

II. THE MODEL

A. Introduction of LR potential

The Hamiltonian of the three-band Hubbard model in the presence of a local repulsion on the copper site U_d and a nearest-neighbour repulsion V reads

$$\begin{aligned} H = &\epsilon_d^0 \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + \epsilon_p^0 \sum_{i\sigma\alpha=x,y} p_{i\alpha\sigma}^\dagger p_{i\alpha\sigma} - t_{pd} \sum_{i\sigma\eta=\pm x,y} sgn(\eta) \left(d_{i\sigma}^\dagger p_{i\eta\sigma} + h.c. \right) \\ &- t_{pp} \sum_{i\sigma} p_{i+x\sigma}^\dagger \left[p_{i-y\sigma} - p_{i+y\sigma} + (i \to i+1) + h.c. \right] + U_d \sum_i n_{i\uparrow}^d n_{i\downarrow}^d + V \sum_{i\sigma\sigma'\eta=\pm x,y} n_{i\sigma}^d n_{i\sigma'}^\eta \end{aligned} \tag{1}$$

Where $d_{i\sigma}^\dagger$ ($d_{i\sigma}$) creates (destroys) a hole in the copper site i, $p_{i\alpha\sigma}^\dagger$ ($p_{i\alpha\sigma}$) in the oxigen site $i+\alpha$, $n_{i\sigma}^d = d_{i\sigma}^\dagger d_{i\sigma}$, $n_{i\sigma}^\eta = p_{i\eta\sigma}^\dagger p_{i\eta\sigma}$ and ϵ_d^0 and ϵ_p^0 are the bare energy levels. We work in hole representation and we denote by δ the number of holes per unit cell added by doping: $\sum_{\sigma,\alpha=x,y} \left[\langle n_{i\sigma}^\alpha \rangle + \langle n_{i\sigma}^d \rangle \right] = (1+\delta)$.

In the case of $U_d = \infty$ (strong-coupling) no double occupancy arises on copper sites $\sum_\sigma d_{i\sigma}^\dagger d_{i\sigma} \leq 1$. We handle this constraint within the standard slave-boson technique [46–50] by introducing a new boson degree of freedom labeling the empty site $d_{i\sigma}^\dagger \to d_{i\sigma}^\dagger b_i, \quad d_{i\sigma} \to b_i^\dagger d_{i\sigma}$. Since the site can only be either singly occupied by a fermion or singly occupied by a boson, the constraint becomes $\sum_\sigma d_{i\sigma}^\dagger d_{i\sigma} + b_i^\dagger b_i = 1$. In order to avoid perturbative assumptions on the coupling constants we use the large-N expansion, assuming the spin index to run from 1 to N and letting $N \to \infty$. So the constraint is relaxed into

$$\sum_\sigma d_{i\sigma}^\dagger d_{i\sigma} + b_i^\dagger b_i = \frac{N}{2}, \tag{2}$$

showing that $b_i \sim \sqrt{N}$. Consistently we rescale the coupling $t_{pd} \to t_{pd}/\sqrt{N}$, $V \to V/N$. Then the partition function of the system can be written as a functional integral

$$\mathcal{Z} = \int \mathcal{D}d_\sigma^\dagger \mathcal{D}d_\sigma \mathcal{D}p_{\alpha\sigma}^\dagger \mathcal{D}p_{\alpha\sigma} \mathcal{D}b^\dagger \mathcal{D}b \mathcal{D}\lambda \mathcal{D}X \mathcal{D}Y \exp\left(- \int_0^\beta d\tau S \right), \tag{3}$$

with

$$S = \sum_{i\sigma} d_{i\sigma}^\dagger \frac{\partial d_{i\sigma}}{\partial \tau} + \sum_{i\sigma\alpha=x,y} p_{i\alpha\sigma}^\dagger \frac{\partial p_{i\alpha\sigma}}{\partial \tau} + \sum_{i\sigma} b_i^\dagger \frac{\partial b_i}{\partial \tau} + i \sum_i \lambda_i \left(b_i^\dagger b_i - q_0 N \right) + \frac{N}{2V} \sum_i \left(X_i^2 + Y_i^2 \right) + \mathcal{H}, \tag{4}$$

$$\begin{aligned} \mathcal{H} = &\sum_{i\sigma} \left(\epsilon_d^0 + i\lambda_i + X_i + iY_i \right) d_{i\sigma}^\dagger d_{i\sigma} + \sum_{i\sigma\alpha=x,y} \left(\epsilon_p^0 - X_i + iY_i \right) p_{i\alpha\sigma}^\dagger p_{i\alpha\sigma} \\ &- \frac{t_{pd}}{\sqrt{N}} \sum_{i\sigma\eta=\pm x,y} sgn(\eta) \left(b_i d_{i\sigma}^\dagger p_{i\eta\sigma} + h.c. \right) - t_{pp} \sum_{i\sigma} p_{i+x\sigma}^\dagger \left[p_{i-y\sigma} - p_{i+y\sigma} + (i \to i+1) + h.c. \right]. \end{aligned} \tag{5}$$

λ is a Lagrange multiplier introduced to enforce the constraint in Eq. (2). Writing the nearest-neighbour Coulomb interaction as

$$\frac{V}{N}\sum_i n_i^d n_i^p = \frac{V}{2N}\sum_i \left[\left(n_i^d + \frac{1}{2}n_i^p\right)^2 - \left(n_i^d - \frac{1}{2}n_i^p\right)^2\right].$$

where $n_i^p = \sum_{i\sigma\eta=\pm x,y} n_{i\sigma}^{\eta}$, $n_i^d = \sum_{i\sigma} n_{i\sigma}^d$, we have decoupled this interaction by means of an Hubbard-Stratanovich transformation introducing the fields X_i and Y_i. Note that in the leading order of the large-N expansion only the Hartree factorization is present, the Fock term being suppressed by a factor $\frac{1}{N}$ due to the sum over the spin. In order to consider the Fock factorization also, one needs to add the term

$$H_{V_2} = \frac{V_2}{N}\sum_{i\sigma\sigma'\eta=\pm x,y} d_{i\sigma}^\dagger d_{i\sigma'} p_{i\eta\sigma'}^\dagger p_{i\eta\sigma}. \tag{6}$$

This term can be decoupled by a complex Hubbard-Stratanovich field $Z_{i\eta}$. However we shall see in Appendix B that the Fock term does not modify the properties of the model from a qualitative point of view. So we discard it from now on.

Now we extend the formalism in order to consider the Coulomb LR potential by taking into account the symmetries of the underling lattice, that is of the square lattice of copper atoms with a two-oxygen basis along the directions $(a/2, 0)$ and $(0, a/2)$. Then we add to the hamiltonian the term:

$$H_{coul} = \frac{1}{2N}\sum_{q\sigma\sigma'} n_{q\sigma}^a \Phi^{ab}(q) n_{q\sigma}^b, \tag{7}$$

where $a, b = d, x, y$ and the sum over a and b is understood.

Φ^{ab} reads as

$$\Phi^{ab}(q) = V_c(q)\begin{pmatrix} 1 & \cos\left(\frac{aq_x}{2}\right) & \cos\left(\frac{aq_y}{2}\right) \\ \cos\left(\frac{aq_x}{2}\right) & 1 & \cos\left(\frac{aq_x}{2}\right)\cos\left(\frac{aq_y}{2}\right) \\ \cos\left(\frac{aq_y}{2}\right) & \cos\left(\frac{aq_x}{2}\right)\cos\left(\frac{aq_y}{2}\right) & 1 \end{pmatrix}, \tag{8}$$

and V_c is given by

$$V_c(q) = \frac{\tilde{V}}{\sqrt{\left\{\frac{\epsilon_\parallel b^2}{\epsilon_\perp a^2}\left[\cos(aq_x) + \cos(aq_y) - 2\right] - 1\right\}^2 - 1}}, \tag{9}$$

We define $\epsilon_\parallel$ and $\epsilon_\perp$ as the dielectric constant in and out the Cu-O plane, a and b the in-plane and out-plane lattice constants; $\tilde{V}$ measures the strength of the Coulomb potential.

$V_c(q)$ is obtained through the solution of the $3D$ Laplace equation for a point-like charge in a cubic lattice, projected onto the $z = 0$ plane [39]. The matrix form of $\Phi^{ab}(q)$ reproduces the structure of the Fourier-space Laplacian operator in the orbital space. In the small q limit $V_c(q)$ has the right behavior

$$\lim_{q\to 0} V_c(q) \sim \frac{1}{q}.$$

corresponding to a two dimensional plane embedded in a three dimensional space.

The partition function including the long-range term now reads

$$\mathcal{Z} = \int \mathcal{D}d_\sigma^\dagger \mathcal{D}d_\sigma \mathcal{D}p_{\alpha\sigma}^\dagger \mathcal{D}p_{\alpha\sigma} \mathcal{D}b^\dagger \mathcal{D}b \mathcal{D}\lambda \mathcal{D}X \mathcal{D}Y \mathcal{D}\Omega^a \exp\left(-\int_0^\beta d\tau \tilde{S}\right), \tag{10}$$

$$\tilde{S} = S + \frac{N}{2}\sum_q \Omega_q^a \Phi_{ab}^{-1}(q)\Omega_q^b + i\sum_{q\sigma}\Omega_q^a n_{q\sigma}^a, \tag{11}$$

where Ω_q^a is a real vector field, introduced to decouple the LR Coulombic interaction. Notice that in Eq. (9) we use the long-wavelength uniform values for the dielectric constants in order to obtain the repulsive Coulomb potential at long-distance. We have also included in the Hamiltonian the explicit n-n Coulomb repulsion V. This term is needed to describe the reduced screening present at short distances between localized charges, which leads to an enhanced repulsion between n-n sites.

B. Large-N Expansion

In this section we present the formalism needed to handle the gaussian fluctuations of the boson fields around the saddle-point solutions. From now on we choose the radial gauge [48]. In this gauge the phase of b_i is gauged away and only the modulus r_i is considered, while λ_i acquires a time dependence. The Hamiltonian of the coupled fermions and bosons is then written in a compact form, by introducing a seven-component field $\mathcal{A}^\mu = (\delta r, \delta\lambda, \delta X, \delta Y, \Omega^a)$ defined as the fluctuating part of the fields around the saddle-point solutions

$$\begin{aligned} r_i &= r_0(1+\delta r_i) \\ \lambda_i &= -i\lambda_0 + \delta\lambda_i \\ X_i &= X_0 + \delta X_i \\ Y_i &= -iY_0 + \delta Y_i \\ \Omega_q^a &= \Omega_q^a. \end{aligned}$$

We have not included the (infinite) zero-momentum component of the Coulombic field Ω_q^a since we are assuming that it is cancelled by the contribution of a uniform charged ionic background.

The Hamiltonian reads

$$\mathcal{H} = \mathcal{H}_{MF} + \mathcal{H}_{bos} + \mathcal{H}_{int}, \tag{12}$$

where, using the notation $\Psi_{k\sigma\alpha} = (d_{k\sigma}, ip_{xk\sigma}, ip_{yk\sigma})$, $\mathcal{H}_{MF}$ is the mean-field fermion hamiltonian (in units of $a = 1$)

$$\mathcal{H}_{MF}(k) = \begin{pmatrix} \epsilon_d & -2r_0 t_{pd}\sin\left(\frac{k_x}{2}\right) & -2r_0 t_{pd}\sin\left(\frac{k_y}{2}\right) \\ -2r_0 t_{pd}\sin\left(\frac{k_x}{2}\right) & \epsilon_p & -2t_{pp}\beta_k \\ -2r_0 t_{pd}\sin\left(\frac{k_y}{2}\right) & -2t_{pp}\beta_k & \epsilon_p \end{pmatrix} \tag{13}$$

where $\epsilon_p = \epsilon_p^0 - X_0 + Y_0$ and $\epsilon_d = \epsilon_d^0 + \lambda_0 + X_0 + Y_0$ are the renormalized energy oxigen and copper levels respectively, and $\beta_k = 2\sin(k_x/2)\sin(k_y/2)$.

The quasiparticle basis $\tilde{\Psi}_k$ is obtained from the unitary transformation, $\tilde{\Psi}_k = U(k)\Psi_k$ which diagonalizes the mean-field Hamiltonian H_{MF}, so that

$$\mathcal{H}_{MF} = \sum_{k\sigma\alpha\beta} \Psi^\dagger_{k\sigma\alpha}\mathcal{H}^{\alpha\beta}_{MF}(k)\Psi_{k\sigma\beta} = \sum_{k\sigma\alpha} E_\alpha(k)\tilde{\Psi}^\dagger_{k\sigma\alpha}\tilde{\Psi}_{k\sigma\alpha}.$$

The boson-fermion interaction term can be written in the form

$$\mathcal{H}_{int} = \sum_{kq\sigma} \Psi^\dagger_{k+(q/2)\sigma}\Lambda^\mu(k,q)\Psi_{k-(q/2)\sigma}\mathcal{A}^\mu(q) = \sum_{kq\sigma} \tilde{\Psi}^\dagger_{k+(q/2)\sigma}\tilde{\Lambda}^\mu(k,q)\tilde{\Psi}_{k-(q/2)\sigma}\mathcal{A}^\mu(q). \tag{14}$$

The 3×3 boson-fermion interaction vertices $\Lambda^\mu(k,q)$ in the orbital operator basis can be obtained from Eq. (11) and are shown in Appendix A. The quasiparticle vertices $\tilde{\Lambda}^{\mu\nu}(k,q)$ are given by

$$\tilde{\Lambda}^\mu(k,q) = U\left(k+\frac{q}{2}\right)\Lambda^\mu(k,q)U^\dagger\left(k-\frac{q}{2}\right). \tag{15}$$

Finally the purely bosonic part of the hamiltonian is

$$\mathcal{H}_{bos} = N\sum_{q\mu\nu} \mathcal{A}^\mu(q)B^{\mu\nu}(q)\mathcal{A}^\nu(-q), \tag{16}$$

where the 7×7 $B(q)$ matrix is given by:

$$B(q) = \begin{pmatrix} B_{sr} & 0 \\ 0 & \frac{1}{2}\Phi^{-1}(q) \end{pmatrix}, \tag{17}$$

and the only nonzero elements of B_{sr} are $B^{11}_{sr} = r_0\lambda_0$, $B^{12}_{sr} = B^{21}_{sr} = ir_0^2$ and $B^{33}_{sr} = B^{44}_{sr} = 1/2V$.
At leading order in $1/N$, the dressed propagator of the $\mathcal{A}^\mu$ field is

$$D^{\mu\nu}(q,\omega) = \langle \mathcal{A}^{\mu}(q,\omega)\mathcal{A}^{\nu}(-q,-\omega)\rangle = \frac{1}{N}\left[2B(q) + \Pi(q,\omega)\right]^{-1}_{\mu\nu}, \tag{18}$$

where $\Pi^{\mu\nu}(q,\omega)$ are the bare polarization bubbles:

$$\Pi^{\mu\nu}(q,\omega) = \sum_{k\alpha\beta} \frac{f_{\alpha}(k+q/2) - f_{\beta}(k-q/2)}{E_{\alpha}(k+q/2) - E_{\beta}(k-q/2) - \omega} \tilde{\Lambda}^{\mu}_{\alpha\beta}(k,q)\tilde{\Lambda}^{\nu}_{\beta\alpha}(k,-q). \tag{19}$$

The factor 2 multiplying the bare boson propagator is due to the fact that in the radial gauge the bosonic fields are real. Correspondingly, at leading order in $1/N$, the density-density correlation functions are given by

$$\chi_{\alpha\beta}(q,\omega) = \frac{1}{N}\sum_{\sigma\sigma'}\langle n_{\alpha\sigma}(q) n_{\beta\sigma'}(-q)\rangle = \chi^{0}_{\alpha\beta}(q,\omega) + \sum_{\mu\nu} P^{0}_{\alpha\mu}(q,\omega) D^{\mu\nu} P^{0}_{\nu\beta}(q,\omega), \tag{20}$$

where

$$\chi^{0}_{\alpha\beta}(q,\omega) = \frac{1}{N}\sum_{\sigma\sigma'}\langle n_{\alpha\sigma}(q) n_{\beta\sigma'}(-q)\rangle_0 \tag{21}$$

are the bare density-density correlation functions, and

$$P^{0}_{\alpha\mu}(q,\omega) = \frac{1}{N}\sum_{\sigma\sigma'}\langle n_{\alpha\sigma}(q) \sum_{k\gamma\delta} \Psi^{\dagger}_{k\sigma'\gamma}\Lambda^{\mu}_{\gamma\delta}(k,q)\Psi_{k+q\sigma'\delta}\rangle_0, \tag{22}$$

with $\alpha = d, p_x, p_y$ and μ running on the boson indices. Linearly combining $\chi_{\alpha\beta}$, we calculate the total density correlation function

$$\chi_{nn}(q,\omega) = \langle (n_p + n_d)(n_p + n_d)\rangle \tag{23}$$

and the charge-transfer correlation function

$$\chi_{CT}(q,\omega) = \langle (n_p - n_d)(n_p - n_d)\rangle. \tag{24}$$

Within this formalism it is possible to calculate the residual (order $1/N$) effective scattering amplitude between quasiparticles in the lowest band (where the Fermi level lays)

$$\Gamma(k,k',\omega) = -\tilde{\Lambda}^{\mu}_{11}(k',-q) D^{\mu\nu}(q,\omega)\tilde{\Lambda}^{\nu}_{11}(k,q). \tag{25}$$

We note in passing that, being the bare bubble defined in Eq. (19) and the vertices defined in Eq. (15) non-singular functions of q, in the present RPA-like approximation the effective interaction Eq. (25) and the correlation functions Eq. (20) always diverge together, since any singularity may only arise from the boson propagator entering both quantities

III. PHASE DIAGRAM AND CORRELATION FUNCTIONS

The three-band extended Hubbard model with short-range interactions has been widely studied in both weak-coupling [43–45,32] and in semianalytic [33,34,36] or fully numeric [51–56] strong-coupling approaches. Before presenting the effects of LR forces on this model, we recall some of the most important results for the pure short-range case. In particular we summarize the phase-diagram in the $\Delta_0 - \delta$ space, $\Delta_0 = \epsilon^0_p - \epsilon^0_d$ being the bare difference between the copper and oxygen atomic levels, for various given values of the nearest-neighbor Cu-O repulsion V in the more realistic strong-coupling approach. At half-filling a metal-charge-transfer-insulator (MCTI) transition is present, for Δ_0 larger than a critical value ($\Delta_0 + V > 3.3 t_{pd}$ when $t_{pp} = 0$). Away from half-filling, increasing V the system shows phase-separation above a critical value V^* ($V^* \simeq 1.63 t_{pd}$ for $t_{pp} = 0$). If V is increased further, two solutions with different occupations of the copper and oxigen orbitals can be found solving the self-consistent equations *at fixed number of particles*. Correspondingly to these solutions, the phase diagram, at finite doping, would display a "first-order" line between a d-like metal and a p-like metal ending in a "second-order" valence instability (VI) point characterized by the softening of the energy of the $p-d$ exciton mode at $q = 0$. However, if the number of particles is allowed to fluctuate the VI line is always inside a region of negative compressibility where the system phase-separates.

Then the VI point is not physically attainable. The region of negative compressibility is delimited by a spinodal line on which the compressibility diverges. Together with the compressibility, all the static charge correlation functions diverge because of the mixing between the charge fluctuations. Along the spinodal line, the exciton mode remains massive (i.e. $\omega_{exc}(q=0) \neq 0$), even if the charge-transfer (CT) correlation function is divergent. The instability is instead accompanied by the overdamped zero sound acquiring a vanishing and then negative velocity. These results are only quantitatively, and not qualitatively, changed if the Fock term in Eq. (6) is considered [34].

When the LR potential of Eq. (7) is added to the short-range extended Hubbard model of Eq. (5), it provides a huge electrostatic cost to the long-wavelength charge fluctuations thereby preventing PS. Nevertheless, an instability region characterized by the divergence of the density-density correlation functions at a finite q_c is still possible and is indeed present, as in the single-band Hubbard-Holstein model, where an ICDW stripe instability was found [5,39]. In Fig. 1 and (2) we show two phase diagrams for two different values of the LR Coulomb force strength $\tilde{V}$: $\tilde{V} = 1.6t_{pd}$ and $\tilde{V} = 8t_{pd}$ with $t_{pp} = 0.2t_{pd}$ and $V = 2.3t_{pd}$ [57]. These values of $\tilde{V}$ would correspond to a repulsion between holes on two nearest-neighbor copper sites of the order $0.1t_{pd}$ and $0.5t_{pd}$) respectively. The phase diagrams are determined by identifying the divergences of the various correlation functions within the $1/N$ expansion at the leading order . The finite-q instability is indicated by the dashed line. When the strength of the Coulomb potential $\tilde{V}$ is larger, as in Fig. 2, the unstable region shrinks around the VI point. The dynamical properties of the extended Hubbard model at $q = 0$ are not affected by the introduction of the LR forces. A remarkable consequence is that the VI point, which can be characterized by $\chi_{CT}(q = 0, \omega \to 0) = \infty$ occurs for the same Δ_0 and δ of the purely short-range case. In the presence of a (large) $\tilde{V}$, part of the first-order VI line lays outside the region of the finite-q instability. However we have checked that the system does not show any critical behavior along this line due to the fact the energy of the exciton-mode remains finite on it and no divergences arise in the response functions as well as in the effective scattering amplitude between quasiparticles.

The static ($\omega = 0$) charge-transfer correlation function $\chi_{CT}(q, \omega = 0)$ is reported in Fig. 3 outside and inside the CDW-unstable region, for $\tilde{V} = 1.6t_{pd}$. Near the instability, a huge peak at $q \simeq q_c$ develops in the charge-transfer as well as in all other correlation functions and they eventually diverge when the charge-instability line is reached.

Although it may be *influenced* by some specific features of the band structure (proximity to a Van Hove singularity, partial nesting of some portion of the Fermi surface and so on), the critical $\mathbf{q_c}$ is not *determined* by any nesting property of the Fermi surface. It rather results from the balance between the tendency towards PS of the short-range model and the electrostatic cost imposed by LR forces to charge segregation. Then its modulus depends both on doping and, less sensitively, on the Coulomb potential through $\tilde{V}$. It increases ranging from 0.5 to 0.8 (in units of the inverse of lattice constant a) with doping and, slightly, with $\tilde{V}$. This behavior reproduces the one already found in the single-band Hubbard-Holstein model, although in this latter case the value of the modulus of q_c was about 20% larger. However, this feature is obviously non-universal and is a specific outcome of the band parameters considered here. We found that the correlation functions are rather isotropic in the (q_x, q_y) space. However a detailed analysis of the direction along which the instability first occurs shows that the $(1, 0)$ and $(0, 1)$ directions are more favorable than the diagonal $(1, 1)$ one.

To clarify the physics underlying the ICDW instability, we studied the behavior of the collective modes at finite q and ω in the stable region. To this purpose, we analyzed the density-density correlation functions [Eqs. (23) and (24)]. The frequency and momentum dependence of the poles (or resonanaces) of the correlation functions as a function of q give indeed the dispersion of the collective modes of the model. Two modes can be identified: The plasmon mode and the charge-transfer exciton. Having introduced a LR term proportional to $1/q$ (for small q) the energy of the plasmon mode vanishes at $q = 0$ as $\sqrt{q}$, whereas the frequency of the exciton mode at $q = 0$ stays finite all over the stable region of the phase diagram. Due to the mixing, both poles are present in all the correlation functions, although with different spectral weight. Whereas the peak of the plasmon is more pronounced in the total density correlation function, the excitonic mode has a larger weight in the CT correlation function.

In Fig. 4 we compare the dispersion of the $2D$-plasmon near and far away the CDW instability. Fig. 4 clearly shows that the $2D$-plasmon mode softens at the same q for which the instability takes place. On the contrary, the exciton mode remains at higher energies and the CDW instability does not affect much its dispersion. We have also analyzed the plasmon mode as a function of the direction of the momentum q in the Brillouin zone. We find that, up to $q \sim 0.5$ (in units of the inverse lattice constant), the dispersion is nearly isotropic and depends very weakly on the direction of q. In particular, when the system is far from the instability, the dispersion on the (1,0) and (1,1) directions is linear (apart from the small region around $q = 0$ where the $q^{1/2}$ behavior is found) with a 10% higher slope in the (1,1) direction. This analysis illustrates that the occurrence of an ICDW instability in the three-band extended Hubbard model is accompanied by the softening of the plasmon mode due to the attractive effective interactions mediated by the charge-transfer fluctuations in the presence of a sizable nearest-neighbor repulsion V. The microscopic dynamics of

the instability is therefore substantially different from the analogous instability taking place in the Hubbard-Holstein model [39]. In this latter case the electronic plasma mode mixed with an optic phonon mode. The finite momentum instability was then accompanied by the softening of the phonon mode, which was also responsible for the effective attraction eventually driving the instability. However, despite the physically different microscopic driving forces and the different dynamical evolution of the collective modes, the similar occurrence of an ICDW instability driven by the interplay of PS tendency and LR forces clearly illustrates the full generality of this mechanism.

IV. EFFECTIVE INTERACTION

Our analysis now proceeds with the investigation of the effective interaction between quasiparticles on the Fermi surface as defined by

$$\Gamma(k_F, k'_F, q; \omega) = -\sum_{\mu,\nu} \tilde{\Lambda}^{\mu}_{11}(k'_F, -q) D^{\mu\nu}(q, \omega) \tilde{\Lambda}^{\nu}_{11}(k_F, q). \tag{26}$$

Fig. 5 displays $\Gamma(q, \omega = 0)$ for $\tilde{V} = 0.8 t_{pd}$ and $\delta = 0.2$, for two different values of Δ_0 close to the CDW instability, as a function of $(q_x, 0)$. As in the case of the short-range model [33–35], near the charge instability, a large attractive interaction between quasiparticles is generated at $\omega = 0$. However, while in the short-range model the interaction diverges negatively at $q = 0$, in the presence of LR forces, $\Gamma(q, \omega = 0)$ stays finite at $q = 0$ and diverge negatively at q_c, in correspondence to the divergence of the correlation functions. This is rather natural since at leading order in $1/N$ the singular behavior of both quantities is related to the singular behavior of the boson propagator.

In the proximity of the instability, $\Gamma(q, \omega)$ can be fitted by the following expression

$$\Gamma(q, \omega) = \tilde{U} - \frac{\hat{V}}{(q - q_c)^2 + \kappa^2 - i\gamma\omega}. \tag{27}$$

$\tilde{U}$ describes the residual, nearly k-independent repulsion mediated by the slave bosons: within the large-N slave-boson formalism, the infinite U repulsion between the bare fermions, is reduced to a rather weak residual repulsion between the Fermi-liquid quasiparticles. The imaginary term in the denominator of Eq. (27) is proportional to ω through the damping coefficient γ and reproduces the behavior of the imaginary part of the bare density-density polarization bubble of the quasiparticles $Im\chi^0_{nn}(\omega, q)$ at small frequencies and finite momenta close to q_c. This means that, despite the complicated structure of the scattering amplitude Eq. (26), in the proximity of the charge instability, the interaction between quasiparticles has a simple RPA-like form

$$\Gamma(q, \omega) = \tilde{U} - \frac{\hat{V}}{1 + \Gamma_\omega(q, \omega)\chi^0_{nn}(q, \omega)} \tag{28}$$

with $\Gamma_\omega(q, \omega)$ being an effective dynamical interaction between the quasiparticles. $\Gamma_\omega(q, \omega)$ is dominantly attractive at $q \sim q_c$ and is mediated by virtual high-energy processes (mostly interband transitions). The behavior of κ^2 as a function of $\delta - \delta_c$ is plotted in Fig. 6. It vanishes as $\alpha(\delta - \delta_c)^{2\nu}$ with $2\nu = 1$. On general grounds, this is what one expects approaching a Gaussian QCP. Since the leading-order $1/N$ expansion bears resemblance with a RPA resummation, it is quite natural that the propagator of the critical fluctuations assumes the universal form of Eq. (27), which is also found in the context of the Hubbard-Holstein model [5,39] and in the proximity of an antiferromagnetic QCP [1], where the instability also occurs for a finite value of $Q_{AF} = (\pi, \pi)$. Of course, the specific form of Eq. (27) could depend from our approximate (nearly mean-field) treatment. Nevertheless the singular nature of the interactions mediated by critical fluctuations is a sound generic consequence. Many physical consequences stem from the presence of a QCP related to an ICDW instability, which have already been generically explored in previous works [5,6]. In particular, the singular interactions were related to an anomalously large decay ratio for the quasiparticles at the "hot" spots $[1/\tau \sim \sqrt{max[\epsilon_k, T]}]$, to linear-in-T resistivity in $2D$ and $\rho \sim T^{3/2}$ in $3D$ [5] and a specific doping dependence of the superconducting critical temperature T_c was found [6]. Similar results are obviously obtained, since the above analyses are quite general in so far they are based on the general theory of QCP's. On the other hand, the peculiar character of the presently considered model obviously enter in the values of the non-universal constants. In particular, whereas the temperature dependence of the mass parameter κ^2 in Eq. (27), can only be calculated within a finite-temperature analysis, other parameters are accessible within our $T = 0$ slave-boson calculation. Specifically, as seen in Fig.6, for typical Hamiltonian parameter, the zero-temperature doping dependence of the mass $\kappa^2 = \alpha(\delta - \delta_c)$ is determined by the value of the coefficient $\alpha \sim 2$ and turns out to be quite smaller than in the phononic Hubbard-Holstein model. As a consequence in the present case the mass term grows more slowly upon increasing the doping away from δ_c, thus leading to a more extended region with substantial critical fluctuations.

V. DISCUSSION AND CONCLUSIONS

The scenario here presented in the context of the three-band Hubbard model involves the relevant interplay between the plasma and the charge-transfer modes. An obvious completion of our work would then regard the possible experimental confirmation of the physical outcomes of the model. Unfortunately, such a connection with experiments is not presently attainable on a quantitative level, due to the lack of detailed experimental analyses on collective electronic excitations at finite momenta.

As far as the limitations of our model are concerned, these are manyfold. First of all, we only considered a purely two-dimensional electron system embedded in a $3D$ inert screening medium, thus neglecting the layered nature of the superconducting cuprates. As a consequence, the investigated collective modes only have a purely two-dimensional character. It is well known that, as far as the $2D$ plasma mode is concerned, it qualitatively differs from the collective plasma modes of a layered electron gas (LEG) [58] formed by an array of $2D$ electron gas layers spaced by a distance d. In particular, the screening induced by the other layers changes the $\sqrt{q}$ dispersion of the $2D$ mode into a continuous branch of modes labelled by the wavevector q_z in the direction perpendicular to the layers, with $-(\pi/d) \leq q_z \leq \pi/d$. Whereas the $k_z = 0$ mode is massive like in $3D$ isotropic systems (plasma oscillations in phase on all different layers), the other modes are acustic with different velocities ($v \to \infty$ for $k_z \to 0$) until at finite momenta in the $2D$ Brillouin zone they all merge in a narrow, infinitely degenerate branch practically indistinguishable from the dispersion of the purely $2D$ mode. However, it is our opinion that some important qualitative indications can already be gained from our simplified model, since the LEG modes merge at $\tilde{q} \sim \pi/d$ substantially smaller than the typical momenta q of the twodimensional Brillouin zone. Therefore, although explicit calculations are still in progress, we believe that the physics at finite sizable momenta of the twodimensional Brillouin zone is not very different for the isolated or the 3D layered systems [59] and a CDW instability is a natural outcome provided q_c is larger than $\tilde{q}$. In our case, we found, $q_c \sim 0.5$ (remember that we take the lattice spacing $a = 1$) thus restricting the validity of our analysis to systems where $\tilde{q} \lesssim 0.5$, i.e. $d > 6a$.

Other important simplifications of the model here considered are worth being investigated and could prevent a direct comparison with real systems. Specifically in the present model, we did not include a small hybridization ($t_\perp$) between adjacent layers, which would render the plasma modes with $k_z \neq 0$ (weakly) massive, thus modifing the dynamics at small momenta. Furthermore the interplay between the mixed collective modes and the particle-hole excitations was considered within a $1/N$ expansion bearing a close resemblance with the RPA approximation. Although extensively used in the literature, this approximation is not guarantied to provide a quantitative description of the physics of the strongly interacting electrons. In particular, at the leading order in $1/N$, both the screening and the damping of the collective modes is determined by the quasiparticles, without any account for the incoherent excitations leading to incoherent particle-hole continua on much larger energy scales. These excitations could in principle affect the dynamics of the system by providing a damping channel for the collective modes even when these latters are outside the quasiparticle-quasihole continuum.

Besides the above limitations of the theoretical model, a comparison with experiments is also made difficult by the lack of detailed experimental analyses of electronic excitations at finite momenta. In particular the extensive wealth of data provided by neutron scattering experiments could be related to the above discussed physical effects only in the case of a strong enough coupling of the CT and plasma collective modes with the lattice or spin degrees of freedom. This could render too indirect the access to the physics of the electronic collective modes, with probably too little intensity to get detectable effects.

On the other hand a direct access to electronic excitations at finite momenta is given by electron-energy-loss spectroscopy. However, the presently available analyses, do not seem to us conclusive in order to confirm or to disprove any anomalous dynamical behavior of the collective plasma modes. In particular, our analysis showed (see Section III) that the anomalous softening of the plasma modes at q_c occurs in a rather narrow range of doping $\delta \sim \delta_c \sim \delta_{opt}$ and momenta $q \sim q_c$. This by itself would require a rather detailed and dedicated search. Moreover, it is to be remembered that (local) superconducting pairing "quenches" the ICDW instability, thereby preventing a complete softening of the plasma mode. Then, presumably the only consequence of the unachieved charge instability would be a partial softening of the mode, likely accompanied by an increase of incoherent spectral weight at low energies near the critical q_c. This latter effect should be separated from the usual substantial background, which likely will render its detection quite a difficult task.

Despite the above theoretical and experimental limitations, the present work provides a definite contribution to the theoretical substantiation of the ICDW-QCP scenario. Indeed we proved that LR coulombic forces do not completely stabilize the tendency towards charge instability in the three-band Hubbard model extended with nearest-neighbour Coulomb repulsion. In particular we explicitely demonstrated the existence of an ICDW instability for a substantial

parameter range of the model and for realistic doping values. Moreover, we showed that, although the VI is still present in the phase diagram and remarkably the excitonic softening at $q = 0$ at the VI critical point is not influenced by the LR forces, the ICDW instability is dominant as it embeds the VI point thus providing the only possible source of critical singular scattering.

Although we have presented the results in the strong-coupling limit, the qualitative behavior of the model does not change in the weak-coupling case (we report the calculations in Appendix B). We have also checked that both in the strong- and the weak-coupling approach the inclusion of a Fock term does not change qualitatively the properties of the system: The CDW region is still present, surrounding the VI point and no critical scattering is found on the VI line.

These results support the idea that incommensurate CDW is a common feature of correlated-electron systems irrespective of the microscopic interaction mechanisms.

Indeed a similar scenario was also found in the framework of one-band Hubbard model with phononic interactions [39]. As far as the collective modes are concerned, their dynamical behavior is obviously related to the specific interactions: In the present case the instability is characterized by the softening of the plasma mode, whereas in the Hubbard-Holstein model the CDW instability is accompanied by the softening of the phonon mode, leading to an instability of the underlying lattice structure. It is quite remarkable that such different dynamics of the collective excitations of the systems, reflecting the underlying microscopic differences of the two models, eventually lead to the same generic conclusion as far as the existence of the ICDW critical point is concerned and the concomitant presence of singular scattering. Indeed, the effective interaction is found in both models to have the same singular behavior as a function of $\delta - \delta_c$ and to display an imaginary part linearly dependent on the frequency. In both models, the singular effective interaction is found to be attractive in the particle-hole channel for a sizeable range of q around q_c in the stable region close to the CDW instability. The scenario presented so far does not include superconductivity. However, it is rather obvious (and it was proven directly in the Hubbard-Holstein model [39]) that the strong effective attraction between quasiparticles, also shows up in the particle-particle channel and leads to (local) Cooper pairing. The occurrence of superconductivity (or of superconducting pairs without long-range coherence) would definitely modify the above scenario in so far it provides an alternative to ICDW. A true long-range CDW order is actually realized overcoming the quenching pairing tendency only when a commensurability condition is realized which pins the charge fluctuations. When commensurability effects do not occur, ICDW fluctuations and superconducting pairing interplay and compete, with strong effects also on the magnetic response of the system. On the one hand ICDW fluctuations create dynamical charge depleted stripes, where magnetic correlations may subsist even near optimal doping. On the other hand the (local) Cooper pairing induced by the singular ICDW scattering can be responsible for the charge- and spin-gap effects which arise in the underdoped cuprates. This complex interplay of ICDW, Cooper pairing and magnetism is not of our concern here, but is surely a most interesting (and difficult) subject which is relevant for the understanding of the cuprates [7,8].

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APPENDIX A: BOSON-FERMION VERTICES

In this section we report the boson-fermion vertices in the orbital basis which are needed in the hamiltonian defined in Eq. (14):

$$\Lambda^1(k,q) = -2r_0 t_{tp} \begin{pmatrix} 0 & \sin\left(\frac{k_x - \frac{q_x}{2}}{2}\right) & \sin\left(\frac{k_y - \frac{q_y}{2}}{2}\right) \\ \sin\left(\frac{k_x + \frac{q_x}{2}}{2}\right) & 0 & 0 \\ \sin\left(\frac{k_y + \frac{q_y}{2}}{2}\right) & 0 & 0 \end{pmatrix}, \tag{A1}$$

$$\Lambda^2(k,q) = \begin{pmatrix} i & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \Lambda^3(k,q) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -\cos\left(\frac{q_x}{2}\right) & 0 \\ 0 & 0 & -\cos\left(\frac{q_y}{2}\right) \end{pmatrix}, \tag{A2}$$

$$\Lambda^4(k,q)=\begin{pmatrix} i & 0 & 0 \\ 0 & i\cos\left(\frac{q_x}{2}\right) & 0 \\ 0 & 0 & i\cos\left(\frac{q_y}{2}\right)\end{pmatrix},\quad \Lambda^5(k,q)=\begin{pmatrix} i & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0\end{pmatrix},\quad \Lambda^6(k,q)=\begin{pmatrix} 0 & 0 & 0 \\ 0 & i & 0 \\ 0 & 0 & 0\end{pmatrix},\quad \Lambda^7(k,q)=\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & i\end{pmatrix}. \tag{A3}$$

APPENDIX B: TWO-BAND MODEL IN WEAK-COUPLING APPROACH

In this Appendix, we present an analytic treatment of a simplified model, showing that (i) the energy of the exciton mode at $q=0$ (denoted by ω_{exc}) is not affected by LR interactions so that the softening of this mode (VI point) takes place in the same point of the phase diagram and that (ii), once the pure SR system is unstable towards phase separation, the system with LR forces always displays a finite-q instability for any value of $\tilde{V}$. In order to keep the formal structure more transparent we confine the calculations to the weak-coupling case [60].

The main difference with respect to the strong-coupling case is given by the absence of the slave-bosons r_q and λ_q needed to treat the infinite on-site repulsion. To further simplify the treatment we also neglect the direct oxigen-oxigen overlap ($t_{pp}=0$). In this case one combination of oxygen orbitals does not hybridize with the copper d orbitals, and gives rise to a flat non-bonding band. Interband processes involving this non-bonding band are decoupled in the small-q limit and, even at finite momenta, do not play any qualitatively relevant role. Therefore, for the sake of simplicity, we will only consider the simplified two-band Hubbard model.

The form factor $\gamma_k=\sqrt{\sin^2(k_x/2)+\sin^2(k_y/2)}$ can be introduced to relate the Fourier transform p_k of the bonding oxygen orbital combination p_i, to the Fourier trasform of the copper d orbitals. The resulting mean-field Hamiltonian is given by:

$$H_{MF}=\sum_{k,\sigma}\mathcal{H}_{MF}(k)=\begin{pmatrix} p_k^\dagger & d_k^\dagger\end{pmatrix}\begin{pmatrix} \epsilon_p & -\sqrt{2}t_{pd}\gamma_k \\ -\sqrt{2}t_{pd}\gamma_k & \epsilon_d\end{pmatrix}\begin{pmatrix} p_k \\ d_k\end{pmatrix}, \tag{B1}$$

where ϵ_d and ϵ_p are the atomic levels, including the Hartree shifts. The mean-field hamiltonian in Eq. (B1) can be diagonalized by a standard unitary transformation, giving the two bands $E_\pm(k)=\left[\epsilon_p+\epsilon_d\pm\sqrt{\Delta^2+8t_{pd}^2\gamma_k^2}\right]/2$. We define $\Delta\equiv\epsilon_p-\epsilon_d$.

We are interested in the analysis of the physical response functions $\hat{\chi}_{\pm,\pm}(q,\omega)$ for the total density (+) and CT (−) fluctuations. A simple analytic progress can only be made in the small-q limit, to which we will confine our treatment here. In this limit the complicated internal structure of the LR interaction [see Eqs. (7)-(9)] greatly simplifies, and the cosine form factors in the nearest-neighbor interaction [see the expressions of the $\Lambda^{3,4}$ vertices in Appendix A] drop. Moreover, in the spirit of weak-coupling approach, we treat the interaction terms in Eq. (1) by means of a standard Hartree decoupling. (We will also show below that adding the contribution of the Fock decoupling does not change qualitatively the behavior of the model).

The Hartree decoupling of the n-n coulomb repulsion is the following

$$V\sum_{i\sigma\sigma'}d_{i\sigma}^\dagger d_{i\sigma}p_{i\sigma'}^\dagger p_{i\sigma'}=V\sum_{i\sigma}\left[2n_i^d p_{i\sigma}^\dagger p_{i\sigma}+2n_i^p d_{i\sigma}^\dagger d_{i\sigma}\right]-8Vn^pn^d \tag{B2}$$

where n_i^d and n_i^p are the local values of the copper and oxygen density per spin.

The resulting interaction matrix (in the total-density and CT basis) reads

$$\hat{V}(q)=\begin{pmatrix} -2V-U/4-V_c(q)/2 & -U/2 \\ -U/2 & 2V-U/4\end{pmatrix}. \tag{B3}$$

with $V_c(q)\approx\tilde{V}/q$ and where the local U interaction on copper was also introduced. At gaussian level, the response functions are given by the matrix relation

$$\hat{\chi}(q,\omega)=-\frac{\hat{\Pi}_0(q,\omega)}{\hat{1}+\hat{\Pi}_0(q,\omega)\hat{V}(q)}. \tag{B4}$$

involving the bare-bubble matrix:

$$\hat{\Pi}_0^{\alpha\beta}(q,\omega) = \sum_{k,\nu} Tr\left\{\hat{\Lambda}^{\alpha}(k+q)\hat{G}^{orb}(k+q,\omega+\nu)\hat{\Lambda}^{\beta}(k)\hat{G}^{orb}(k,\nu)\right\}, \tag{B5}$$

where $\hat{G}^{orb}_{\alpha,\beta}(k,\nu)$ is the matrix of the Fermionic Green functions in the orbital basis ($\alpha,\beta = d,p$) and $\hat{\Lambda}^{\alpha}(k)$ are the vertices, which couple the p and d fermions to the density fluctuations

$$\hat{\Lambda}^{+}(k) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \hat{\Lambda}^{-}(k) = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{B6}$$

The value of the CT exciton frequency at zero momentum can be extracted from the pole of the CT correlation function, since, due to particle conservation, the total density (+) decouples from the dynamics. This shows up in the $q \to 0$, $\omega \neq 0$ behavior of the bubbles containing the total-density vertex:

$$\lim_{q\to 0} \Pi_0^{+\alpha}(q,\omega) \sim q^2, \tag{B7}$$

($\alpha = +,-$). Then, since $V_c(q) \sim \tilde{V}/q$ always couples to a total-density vertex of $\Pi_0^{+\alpha}$, the diverging Coulomb potential is cancelled by the small-q behavior of the bare bubbles having at least one (+) vertex and does not appear in the $q = 0$ limit of Eq. (B4). By introducing the notation

$$\Pi^{\mu\nu}_{0\omega} = \lim_{q\to 0} \Pi_0^{\mu\nu}(q,\omega) \qquad \Pi^{\mu\nu}_{0q} = \lim_{\omega\to 0} \Pi_0^{\mu\nu}(q \simeq 0,\omega),$$

one obtains

$$\chi_{CT}(0,\omega) = \chi_{--}(0,\omega) = \frac{\Pi^{--}_{0\omega}}{\omega^2 - \left(1 + \Pi^{--}_{0\omega} V_{--}\right)}. \tag{B8}$$

Notice that V_c no longer appears in Eq. (B8), thus showing that $\omega_{exc}(q=0) = \left(1 + \Pi^{--}_{0\omega} V_{--}\right)^{1/2}$ is the same as in the pure SR case.

In the opposite static limit $\omega = 0$ and small q

$$\chi_{--}(q) = \chi_{CT}(q) = -\frac{\left\{\Pi^{++}_{0q}\Pi^{--}_{0q} - \left(\Pi^{+-}_{0q}\right)^2\right\} V^{++}}{\left\{\Pi^{++}_{0q} + V^{--}\left[\Pi^{++}_{0q}\Pi^{--}_{0q} - \left(\Pi^{+-}_{0q}\right)^2\right]\right\} V^{++} + A_H^2}. \tag{B9}$$

with

$$A_H^2 = 1 + V^{--}\Pi^{--}_{0q} + 2V^{+-}\Pi^{+-}_{0q} + \left(V^{+-}\right)^2\left[\left(\Pi^{+-}_{0q}\right)^2 - \Pi^{++}_{0q}\Pi^{--}_{0q}\right] > 0. \tag{B10}$$

Using the relation

$$\lim_{\omega\to 0} \Pi^{--}_{0\omega} = \lim_{q\to 0}\left[\Pi^{--}_{0q} - \frac{\left(\Pi^{+-}_{0q}\right)^2}{\Pi^{++}_{0q}}\right] \tag{B11}$$

and noting that, for small q's, V^{++} is dominated by the LR coulombic term $\tilde{V}/q$, a simpler form can be obtained for $\chi_{--}(q)$

$$\chi_{--}(q) \approx \frac{\tilde{V}\Pi^{--}_{0\omega}}{\left(\omega^2_{exc} + B^2q^2\right)\tilde{V} + q\left(A_H^2/\Pi^{++}_{0q}\right)} \tag{B12}$$

It worth noting that, due to this relations, the static limit of the correlation function equals the dynamical limit, provided ω_{exc} is finite. This fact depends on the presence of the LR forces: in the dynamical limit the intra-band processes are switched off and only inter-band transition are allowed; in the static limit both processes are allowed, but the LR interactions drastically reduces the intra-band transitions at $q = 0$, while it allows the inter-band ones. This also justifies why the exciton energy does not change introducing LR forces: ω_{exc} is indeed involved in inter-band processes which are not affected.

The zeros of the denominator of Eq. (B12) can be obtained through the equation

$$\tilde{V}\left(\omega_{exc}^2 + B^2q^2\right) - A^2q = 0. \tag{B13}$$

with $A^2 \equiv A_H^2/\Pi_{0q}^{++}$. If $\omega_{exc} < A^2/B\tilde{V}$, Eq. (B13) has two real solutions showing the instaurance of a finite-q instability

$$q_1 \simeq \frac{\omega_{exc}^2\tilde{V}^2}{A^2} \qquad q_2 \simeq \frac{A^2}{\tilde{V}B^2}, \tag{B14}$$

Approaching the VI point upon changing the doping, ω_{exc} decrease. Eventually it reaches the *finite* critical value ω_{exc}^c where the charge-transfer (and any other) correlation function diverges at finite-q and the instability takes place. From the above result we see that there is a direct connection between the lowering of the exciton energy ω_{exc} and the developing of a region of instability with the finite-q softening of the plasmon mode. A similar behavior can be found in the SR model [33], where the $q = 0$ instability is driven by the partial softening of the exciton mode which pushes the zero-sound into the continuum eventually leading the system to a phase-separation instability.

Finally, we show that the inclusion of a Fock contribution does not change the qualitative behavior of the model. For the sake of simplicity we limit ourself to the weak-coupling approach, but the same conclusions hold also in the strong-coupling case. In this case we have to add to Eq. (B2) the following terms:

$$V\sum_{i\sigma}\left(Z_i d_{i\sigma}^\dagger p_{i\sigma} + Z_i^\dagger p_{i\sigma}^\dagger d_{i\sigma}\right) - 2VZ^\dagger Z \tag{B15}$$

We introduce two suitable linear combinations of the Fock bosons:

$$A_i = \frac{1}{2}\left(Z_i + Z_i^\dagger\right)$$
$$B_i = \frac{i}{2}\left(Z_i - Z_i^\dagger\right)$$

Considering a mean-field and fluctuating part for these bosons ($A_i = A_0(1+\delta A_i)$, $B_i = B_0(1+\delta B_i)$), two new vertex are needed in the bare-density-correlation functions (we note in passing that the A-vertex has the same structure of the r-vertex in the strong-coupling approach) :

$$\hat{\Lambda}^A(k) = \begin{pmatrix} 0 & -2VA_0\gamma_k \\ -2VA_0\gamma_k & 0 \end{pmatrix}, \quad \hat{\Lambda}^B(k) = \begin{pmatrix} 0 & 2VB_0\gamma_k \\ -2VB_0\gamma_k & 0 \end{pmatrix}. \tag{B16}$$

Due to the fact that the boson B_i decouples from the others we can easily write the dynamical limit of the correlation function $\chi_{CT}(q)$ including the Fock terms:

$$\chi_{CT}(\omega) = \chi_{--}(\omega) = \frac{\Pi_{0\omega}^{--}}{\omega^2 - \left(1 + \Pi_{0\omega}^{--}V_{--} + \Pi_{0\omega}^{AA}V_{AA}\right)}, \tag{B17}$$

where $V_{AA} = -2V$. This result is the same as the one obtained without the Fock term apart from a contribution proportional to V_{AA} which simply renormalizes the exciton-mode energy ω_{exc}^{HF} [34].

Furthermore the statical limit is:

$$\chi_{--}(q) = \chi_{CT}(q) = -\frac{\left\{\Pi_{0q}^{++}\Pi_{0q}^{--} - \left(\Pi_{0q}^{+-}\right)^2\right\}V^{++}}{\left\{\Pi_{0q}^{++} + V^{--}\left[\Pi_{0q}^{++}\Pi_{0q}^{--} - \left(\Pi_{0q}^{+-}\right)^2\right] + V_{AA}\left[\Pi_{0q}^{++}\Pi_{0q}^{AA} - \left(\Pi_{0q}^{+A}\right)^2\right]\right\}V^{++} + A_{HF}^2}. \tag{B18}$$

This expression can be cast in the same form of Eq. (B9) provided ω_{exc} is replaced by ω_{exc}^{HF} and so all the conclusions we have drawn after Eq. (B12) still hold.

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Figure Captions

Fig. 1: Phase diagram in the plane $\epsilon_p^o - \epsilon_d^o$ vs δ for $t_{pp} = 0.2t_{pd}$, $V = 2.3t_{pd}$. The Coulomb interaction strenght is taken so that the repulsion between two nearest-neighbor copper atoms, $V_c(d-d)$, is $0.1t_{pd}$.

Fig. 2: Phase diagram in the plane $\epsilon_p^o - \epsilon_d^o$ vs δ for $t_{pp} = 0.2t_{pd}$, $V = 2.3t_{pd}$. The Coulomb interaction strenght is taken so that the repulsion between two nearest-neighbor copper atoms, $V_c(d-d)$, is $0.5t_{pd}$.

Fig. 3: Charge-Transfer correlation function $\chi_{CT}(q,0)$ vs momentum for $\delta = 0.12$, $\tilde{V} = 1.6t_{pd}$ and $\epsilon_p^o - \epsilon_d^o = 1.835t_{pd}$ $(+)$, $\epsilon_p^o - \epsilon_d^o = 1.84t_{pd}$ $(\Box)$, $\epsilon_p^o - \epsilon_d^o = 1.845t_{pd}$ $(\times)$, $\epsilon_p^o - \epsilon_d^o = 1.85t_{pd}$ $(\diamond)$. The critical value is $\epsilon_p^o - \epsilon_d^o \sim 1.85t_{pd}$.

Fig. 4: Plasma frequency ω_{pl} vs momentum for $\delta = 0.12$ and $\epsilon_p^o - \epsilon_d^o = 1.83t_{pd}$ $(\diamond)$, $\epsilon_p^o - \epsilon_d^o = 1.845t_{pd}$ $(+)$; the critical value is $\epsilon_p^o - \epsilon_d^o \sim 1.85t_{pd}$

Fig. 5: Effective Interaction $\Gamma(q,0)$ vs momentum for $\delta = 0.2$ and $\epsilon_p^o - \epsilon_d^o = 1.92t_{pd}$ $(\diamond)$, $\epsilon_p^o - \epsilon_d^o = 1.922t_{pd}$ $(+)$. The CDW instability corresponds to $\epsilon_p^o - \epsilon_d^o \sim 1.923t_{pd}$

Fig. 6: Doping dependence of the mass parameter at $T = 0$ near the critical point.